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Original Article

Eco-friendly recycled polypropylene matrix composites incorporated with geopolymer concrete waste particles

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ABSTRACT

Civil construction wastes have been incorporated into polymers for recycling as novel engineering composites. In the present work eco-friendly composites with recycled polypropylene (rPP) matrix incorporated with geopolymer concrete waste particles, wither plain (GCW) or surface-modified with oleic acid (AGC) were investigated. The geopolymer concrete waste particles were mixed with polymer powder to provide an effective dispersion between the different materials. Composites were produced by an initial reactive extrusion processing followed by injection molding. These novel composites with amount of 20, 40 and 50 wt% of GCW particles, both plain as-received and surface-modified, were technically evaluated by tensile tests, statistically analyzed by ANOVA, as well as by water absorption as per ASTM standards. Surface dispersion of nanoparticles was revealed by atomic force microscopy. Microstructural analysis was performed by scanning electron microscopy. The results indicated that these sustainable GCW particles incorporated into rPP matrix exhibit superior processability and water absorption less than 0.01%. The rPP/AGC composites present relatively higher elastic modulus, 629 MPa, as compared to the neat rPP, with 529 MPa. These properties suggest potential sustainable applications in building construction using waste materials.

1. Introduction

The construction industry is, in past years, investigating innovative materials to substitute traditional ones, such as wood, polymers, concrete and metals [1]. In particular, polymer matrix composites are replacing conventional materials owing to intrinsic characteristics such as low density and plasticity. Among these composite those reinforced with natural fibers compose a class of successful materials increasingly used in building constructions [2–5]. It is worth mentioning that, within this class, nanocellulose is a promising reinforcement for polymer composites with special proprieties [6–17]. In addition to natural fibers, wastes from building construction incorporated into polymer matrices constitute another important class of eco-friendly composites with specific recycling purpose [18,19].

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In terms of thermoplastics, polypropylene (PP) is one of the most important polymers, produced in large scale and commonly applied at several sectors, including textile, automotive, laboratory equipment, plastic parts and reusable containers. However, its huge post-consumed volume has generated worldwide large amounts of urban solid residues. On the other hand, important characteristics such as high chemical stability, stiffness, hardness, toughness, heat resistance and mechanical properties still remain in spent PP, making it an attractive recycled waste to be use as high performance composite matrix [20–23]. Indeed, several works investigated the application of inorganic fillers in PP matrix composites. Sha et al. [24] investigated innovative methods of filler modification in the production of thermoplastic composites enabling the increase of the strength, stability of interface bonding, high dispersion of particles and appropriated rheology. Pedrazzoli and Pagoretti [25] reported on the properties of PP composites reinforced with expanded graphite nanoplatelets. Zhai et al. [26] investigated the recycling asbestos tailings used as fillers in PP composites. Ahmeda et al. [27] investigated the influence of filler loading on the properties of polypropylene/marble sludge composites and reported that the performance of the polymer composites depends on the relationship between the interfaces of the matrix and fillers. Chen et al. [28] studied the solubility and diffusivity of CO_2 in polypropylene/micro-calcium carbonate composites. The effects of the fillers and interface bonding condition between the fillers and polymer matrix were reported by the authors. Etcheverry et al. [29] studied the effect of adhesion and mechanical properties through chemical anchorage of polypropylene onto glass fibers, previously treated with methylaluminoxane. Zhu et al. [30] observed the influence of the montmorillonite on mechanical properties, crystallization and rheological behaviors of PP composites.

An innovative material developed to replace conventional cement in building construction is the geopolymer, produced from a chemical reaction of silica (SiO_2) and alumina (Al_2O_3) to form an inorganic polymer by geopolymerization [31–33]. In fact, geopolymers have gained considerable attention owing to their intrinsic characteristics such as fire resistance, high toughness and lower CO_2 emissions during productive circle, as compared to Portland cement [34]. Geopolymers can also be used in composite materials. Ferdous et al. [35] observed the influence of geopolymer fillers in the production of hybrid composites, such as the red mud, fly ash and asbestos tailings. As a civil construction cement, a geopolymers might be incorporated with additives such as sand and pebbles to make concrete [36]. During construction and after demolition, geopolymer concretes may become wastes and thus could be recycled by addition as a second phase to a polymer matrix. A similar situation may also occur with a PP product, which after operational life becomes a waste. In this case, the easy to mold thermoplastic PP could be recycled as composite matrix. Based on the aforementioned results, the objective of this work was to evaluate the processability and the tensile behavior as well as to perform a microstructural analysis of eco-friendly composites with incorporation of oleic-acid modified geopolymer concrete (AGC) waste particles into the recycled polypropylene (rPP). For the first time AGC waste is incorporated into a recycled polymer to develop a sustainable novel composite.

2. Materials and methods

2.1. Materials

The plain geopolymer concrete waste (GCW), an as-received material, was supplied by Lafarge Concreto Ltda., Brazil. Recycled polypropylene (rPP), from products molded by blow processing was supplied by COMBRARE Comercial Brasileira de Reciclagem Ltda. The oleic acid (99% purity) was supplied by Sigma Aldrich, Brasil. Recycled PP composite incorporated with both plain as-received GCW particles and oleic acid surface modified (AGC) particles were investigated.

2.2. Materials

The as-received GCW was crushed in particles smaller than 270 mesh and treated with oleic acid during 24 h, as reported elsewhere [18]. The modified particles of AGC were dispersed into molten rPP matrix in a screw extruder. The temperature profile, starting from the feeding zone up to the die, was 180, 190 and 200 °C, while the screw rotating rate was maintained at 6 rpm [37]. The proportions produced of rPP/AGC correspond to 80/20, 60/40, 50/50 (m/m%). Pellets of the modified composites were processed in a Battenfeld injection machine, Plus 35 model. The blend mixtures were performed at 200 °C. According to ASTM C272 [38] and ASTM D638 [39] standards, water absorption and tensile properties were evaluated, respectively. The influence of the modifier fillers was observed during the processing of the AGC specimens, showing greater fluidity and low torque, as well as better molding and processability for the AGC composites, independently of the amount processed. Instead of what was expected, a higher fluidity was acquired by increasing the proportion of modified geopolymer concrete into the polymer matrix of the AGC composites. This effect can be explained by the plasticizing influence of the oleic acid fixed on the surface of the hybrid fillers, while was blended into the rPP matrix [40].

Atomic force microscopy (AFM) was performed to evaluate the agglomerated and surface condition of the polymer composites. Observation was carried on a XE7 equipment, produced by Park System.

Water absorption evaluation as per ASTM C272 [38] was performed to evaluate the percentage of water absorption in the rPP as well as in both rPP/GCW and rPP/AGC composites, considering the results of seven specimens for each.

Tensile tests as per ASTM D638 [39], were carried out on a universal model 1185 Instron machine. The elastic modulus (between 0.05% and 0.25% strains) at a crosshead speed of 5.105 mm/min, and tensile strength at the same crosshead speed, of the rPP/AGC composites were calculated. Analysis of variance (ANOVA) was performed for statistical validation of the properties obtained from ten tensile-tested specimens of each type of composites, including the rPP (0% AGC).

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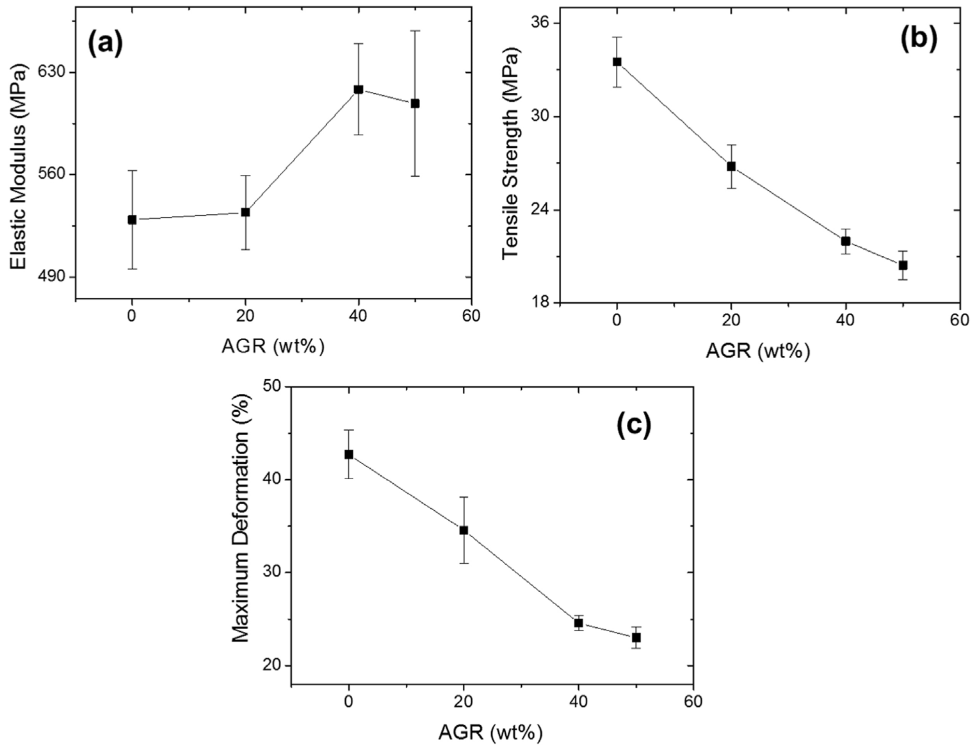

Fig. 1 – (a) Elastic modulus; (b) tensile strength, and (c) maximum deformation variation with recycled polypropylene (rPP) composites incorporated with oleic acid modified geopolymer concrete waste (AGW).

Table 1 – Water absorption (wt%) of injection molded recycled polypropylene (rPP) composites incorporated with plain geopolymer concrete waste (GCW) and oleic acid modified geopolymer concrete waste (AGC).

Fraction of AGC Incorporation (wt%)	rPP/GCW	rPP/AGC
0	0.01	≤0.01
20	≤0.01	≤0.01
40	≤0.01	≤0.01
50	≤0.01	≤0.01

3. Results and discussion

Table 1 presents the results of water absorption tests for pure rPP, 0% geopolymer concrete waste, as well as for both rPP/GCW and rPP/AGC composites. According to these results, all investigated materials absorbed practically no amount (≤0.01%) of water, what corroborates previous results [18]. They also indicate no significant effect of capillary water absorption by geopolymer concrete incorporated into the PP matrix, which reveals consistently embedded plain GCW and modified AGC into rPP.

The tensile properties of the rPP/AGC composite specimens and the rPP matrix (0% AGC) are presented in Table 2. The observed results show the influence of the high amount of modified AGC fillers into the matrix. Tensile results for the rPP/GCW composites were less significant for this present investigation and, therefore, are not presented herein. The good interaction between modified AGC and rPP enables the increase of the elastic modulus of the composites by the incorporation of modified geopolymer concrete waste particles. Furthermore, the results show that the rPP matrix decrease the maximum deformation from 42.7% to about 24%, stabilizing this level at the 40–50% AGC proportions. The increase of the elastic modulus is an indicative of a good adhesion between matrix/fillers. Consequently, the composites acquired suitable stiffness capacity for civil construction applications. These results corroborates those already obtained for PP incorporated industrial wastes [26,27,29].

Fig. 1 shows the graphs corresponding to the results presented in Table 2. As for the variation of the elastic mod-

Table 2 – Tensile properties of recycled polypropylene (rPP) composites incorporated with oleic acid modified geopolymer concrete waste (AGC).

Fraction of AGC incorporation (wt%)	Tensile Strength (MPa)	Elastic Modulus (MPa)	Maximum Deformation (%)
0	33.5 ± 1.6	529.1 ± 33.9	42.7 ± 2.6
20	26.8 ± 1.4	534.1 ± 25.3	34.6 ± 3.6
40	22.0 ± 0.8	618.2 ± 31.2	24.6 ± 0.8
50	20.4 ± 0.9	608.6 ± 49.8	23.0 ± 1.2

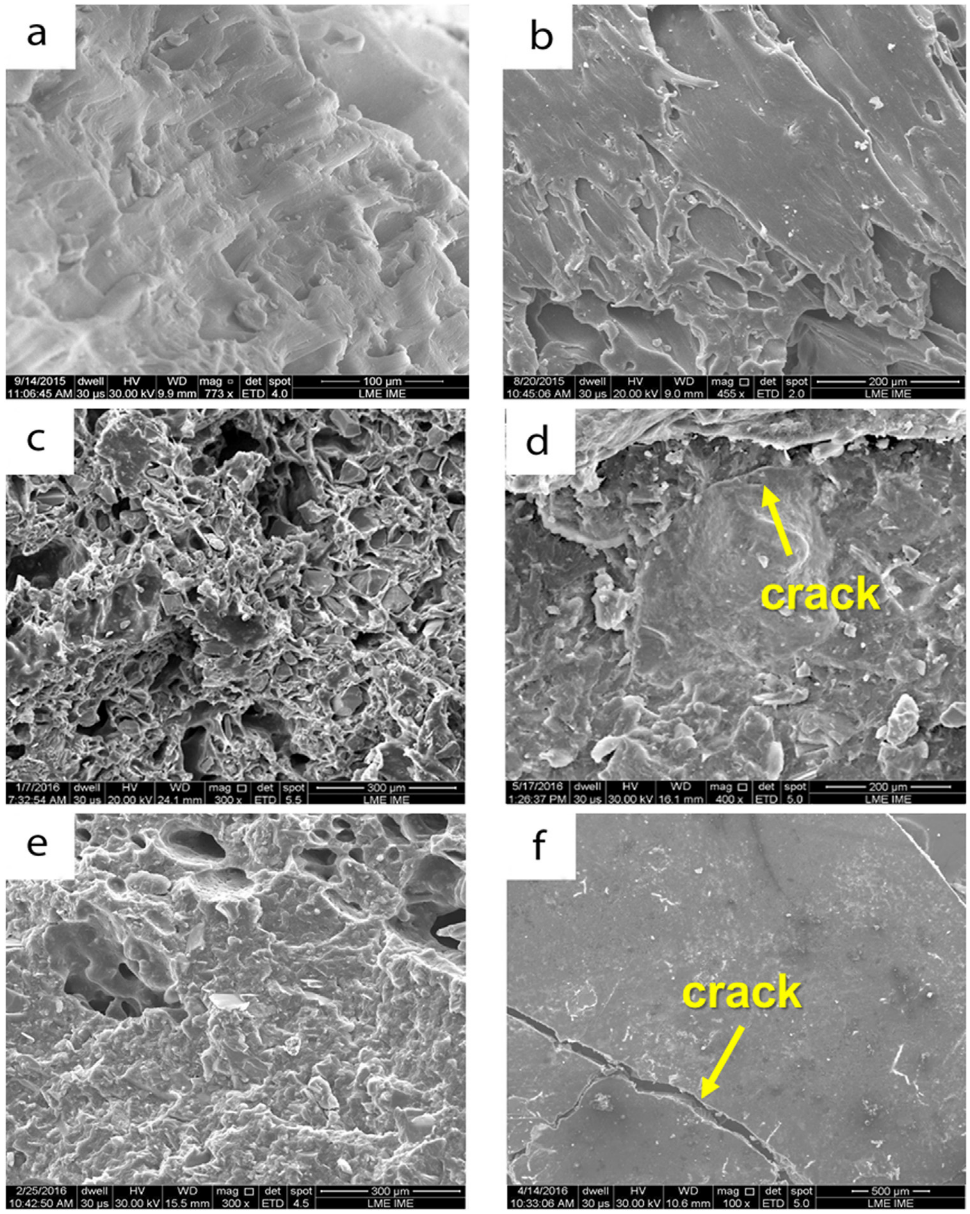

Fig. 2 – SEM fractography of recycled polypropylene (rPP) composites incorporated with oleic acid modified geopolymer concrete waste (AGC) in conditions of previous extrusion (a), (c) (e); and final injection molded (b), (d), (f).

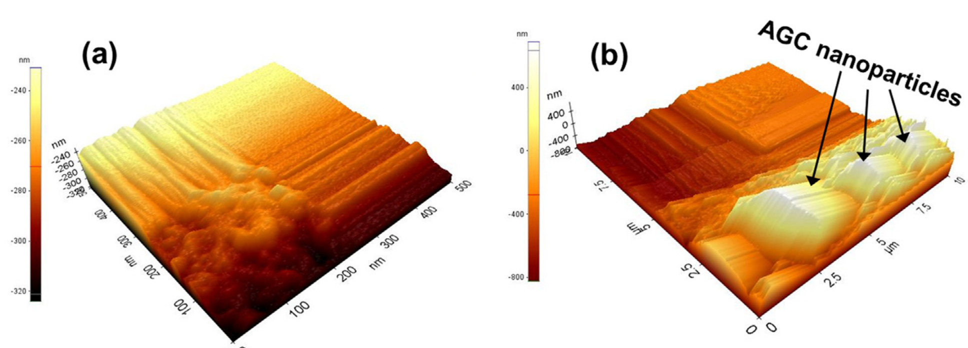

Fig. 3 – Atomic force microscopy of recycled polypropylene (rPP) composites incorporated with: (a) 20 wt% and (b) 50 wt% of oleic acid modified geopolymer concrete waste (AGC).

Table 3 – Analysis of variance (ANOVA) parameters for tensile properties of recycled polypropylene (rPP) composites incorporated with oleic acid modified geopolymer concrete waste (AGC).

Anova Parameters	F	Fc	P
Tensile Strength	161.32	3.01	4.88×10^{-16}
Elastic Modulus	12.04	3.01	5.25×10^{-5}
Maximum Deformation	109.18	3.01	4.00×10^{-14}

ulus, Fig. 1(a) reveals a significant increase in the elastic modulus above 20% incorporation of oleic acid surface treated-geopolymer concrete waste. This corresponds to an effective reinforcement in the stiffness of rPP/AGC composites. By contrast, the tensile strength, Fig. 1(b), and maximum (total) deformation, Fig. 1(c), suffer a decrease with AGC incorporation. The reason for these adverse results in the strength and deformation might be attributed to generation of cracks as further discussed regarding the microstructural evolution.

The data presented in Table 2 and shown in Fig. 1 were validated by ANOVA. Table 3 presents the values of the ANOVA parameters F, F critical (Fc) and P. In this table, considering that 5% (0.05) is the level of significance, values of $F > Fc$ are associated with 95% of confidence in the difference between them. Moreover, if $P < 0.05$, a significant difference might be considered between values.

As shown in Table 3, based on the ANOVA, the incorporation of modified AGC into rPP causes a statistically significant increase in the elastic modulus. In other words, the hypothesis that the values are the same can be rejected with 95% of confidence.

Fig. 2 shows SEM fractographs of 20, 40 and 50% AGC incorporated into rPP composites, each one in both processing conditions of initially extruded, Fig. 2(a), (c) and (e), and finally molded by injection, Fig. 2(b), (d) and (f). The initially extruded condition reveals a fracture associated with more pores then the finally injected. This clearly indicates that the second stage of injection processing has significantly decreased the porosity, mainly for 50% AGC composite in going from extruded condition, Fig. 2(e), to mold-injected, Fig. 2(f). It confirms an effective encapsulating of the geopolymer concrete waste particles by the recycled polypropylene matrix. This porosity elimination after injection corroborates the practically null water absorption results in Table 1. On the other hand, the injected specimens display evidence of cracks as pointed for the 40 and 50% AGC composites in Fig. 2(d) and (f), respectively. This cracking observation could explain the decrease in strength and total deformation in Table 2 and Fig. 1.

Fig. 3 shows AFM results for two investigated extreme conditions of rPP composites, with 20 and 50% AGC. This figure evidences the change in roughness, from a relatively smother surface for the 20% AGC composite in Fig. 3(a) to a rougher surface for the 50% AGC in Fig. 3(b). Actually, the cause of surface roughness for the 50% AGC composite might be assigned to the agglomeration of oleic acid modified geopolymer concrete waste nanoparticles, as pointed by arrows in Fig. 3(b). The results in Fig. 3 disclose the significant influence that AGC particles promote in the composite surface topography. As discussed, the results in Fig. 2 for the 50% AGC composite indicate an effective encapsulating condition, which should be interpreted as good adhesion between particles and rPP matrix. This is apparently confirmed by the well matrix-embedded AGC particles pointed in Fig. 3(b), which certainly favors a stiffness reinforcement presented in Table 2 and depicted in Fig. 1(a).

4. Summary and conclusions

- Eco-friendly composites with recycled polypropylene (rPP) matrix incorporated with 20, 40 and 50 wt% of geopolymers concrete waste particles, both plain as-received (GCW) and oleic acid surface-treated (AGC) were for the first time processed and characterized.
- An initial extrusion followed by injection molding result in rPP/AGC composites displaying greater fluidity in association with low extrusion torque as well as improved molding if compared with rPP/GCW composites.
- Practically no water absorption (≤0.01%) was found for both types of rPP/GCW and rPP/AGC composites, including for pure rPP.
- The incorporation of AGC particles above 20 wt% significantly increases the elastic modulus of the rPP matrix. However, this incorporation reduces the tensile strength and total strain.
- SEM fratographs revealed an effective reduction of porosity during processing from extrusion to injection molding, which justify the null water absorption. However, cracking after molding would be responsible for decrease in strength and strain.
- AFM discloses evidence of rougher surface due to particles agglomeration in composite with high amount of AGC, which might favor the obtained stiffness reinforcement.

Conflicts of interest

The authors declare no conflicts of interest.

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